“Return to the Soil” Nanopaper Sensor Device for Hyperdense Sensor Networks

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Abstract

A nanopaper sensor device that combines humidity sensing, wireless information transmission, and degradability has been fabricated using wood-derived nanopaper as the substrate and dielectric layers. The nanopaper shows excellent suitability for capacitor dielectric layers because of its high dielectric constant, insulating properties suitable for thin film formation, and lamination properties. A wireless transmission circuit containing the nanopaper capacitor can transmit radio signals in the megahertz band, and the relative humidity change can be output as a change in the radio signal owing to the humidity sensitivity of the nanopaper capacitor. More than 95% of the total volume of the nanopaper sensor device decomposes in soil after 40 days. Because the nanopaper sensor device does not need to be recovered, it is expected to greatly contribute to a sustainable society through realization of hyperdense observation networks by mass installation of sensor devices.

1. Introduction

In recent years, wireless sensor networks consisting of a lot of environmental sensing devices have played an important role in manufacturing, agriculture, weather forecasting, management of energy consumption, and so forth.[1–4] Proliferation of sensor devices and internet of things (IoT) enables valuable information to be collected from everywhere and enhances the way we live and work by introduction of a wealth of new services. However, the rapid increase of sensor devices is accompanied by the necessity of enormous maintenance and disposal, which hinders development of a smart society. Moreover, it is difficult to collect all of the sensor devices installed in large quantities over a wide area, for example, for disaster prediction or smart agriculture.[1,4] This means that uncollected sensor devices, IoT waste, may cause environmental pollution.[5–7] To solve the bottleneck of the IoT, there is a need for "degradable" sensor devices that almost completely decompose without any residue or hazardous material after use.

Several studies have reported degradable elements, such as sensors, transistors, conductive lines, and substrates.[8-10] However, it is difficult to combine the functions required for sensor devices and degradability. Degradable sensor devices require three functions: wireless communication, sensing, and degradability. Although the sensor characteristics and degradability of degradable elements have been discussed in other studies,[10] sensor information collection currently requires wired communication with non-degradable external equipment. Sensors output environmental changes, such as temperature, humidity and strain changes, as resistance and capacitance changes. The changes from sensors are basically weak, and signal processing circuits consisting of numerous elements, including resistors, coils, and capacitors, are essential to

amplify and convert the signals for wired/wireless transmission. This is the same for degradable sensor devices. Degradable sensor devices should include degradable sensors and degradable signal-processing circuits. In other words, to realize degradable sensor devices, the substrate, sensor, and passive elements constituting the device should have degradability and sufficient performance, and it is necessary to connect and mount them using an existing and eco-friendly method without impairing the degradability. To achieve this difficult challenge, a smart green material that combines heat resistance, degradability, and suitability for multiple purposes, such as substrates, dielectric layers, and sensors, is required.

Nanopaper, a transparent film consisting of 3–15 nm wide cellulose nanofibers, has attracted attention as a substrate for flexible electronic devices.[11–21] Owing to the high thermal stability, high surface smoothness, and flexibility of nanopaper, flexible electronics based on nanopaper have been developed, including transparent electrodes, organic solar cells, transistors, antennas, and memory devices.[12–18] In addition, nanopaper is biodegradable, similar to conventional paper,[17] so nanopaper is a promising carbon-neutral substrate for degradable sensor devices. The performance of nanopaper as a substrate has been discussed in many studies,[12-18] however, its use as a dielectric layers and sensors has not been fully discussed. Moreover, there is a challenge regarding the integration of nanopaper elements.

Herein, we report a nanopaper sensor device for environmental monitoring that has the three functions of wireless communication, sensing, and degradability using nanopaper as the substrate, dielectric layer, and sensor. The suitability of the nanopaper for capacitor dielectric layers has been demonstrated, and the thermal resistance, low thermal expansion, and printability of the nanopaper have allowed all elements to be successfully integrated to realize a nanopaper wireless transmitter. The flexible and degradable sensor device is capable of humidity sensing

due to the humidity sensitivity of the nanopaper, wirelessly transmitting the data, and degrading in soil after use.

2. Experimental Section

2.1. Cellulose Pulp and Nanofibrillation

Holocellulose pulp was prepared by the steps described in a previous study [21]. First, 60 g of Japanese cedar (Cryptomeria japonica) wood chips were dewaxed in a mixture of acetone/water (2700 mL/300 mL) overnight at room temperature with gentle stirring. The chips were then delignified in an acetic anhydride/hydrogen peroxide mixture (500 mL/500 mL) at 90 °C for 2 h. Next, the holocellulose pulp was treated with 5 wt % potassium hydroxide at 20 °C for 2 h before being thoroughly washed with distilled water. Disintegration of the cellulose pulp into nanofibers was performed using a water-jet nanofibrillation system [15,21]. The pulp slurry (2000 g, pulp content 0.50 wt %) was homogenized using a high-pressure water-jet system (Star Burst, HJP-25008, Sugino Machine Co., Ltd., Toyama, Japan) equipped with a ball-collision chamber. The injected slurry was repeatedly passed through a small nozzle with a 0.15 mm diameter under pressure of 245 MPa. After 50 passes through this nozzle, a 0.32 wt % cellulose nanofiber water dispersion was obtained.

2.2. Conditioning of the Cellulose Nanofiber Dispersion

The concentration of the cellulose nanofiber dispersion was adjusted by condensation with a rotary evaporator (EYLA SB1200, Tokyo Rikakikai Corp., Tokyo, Japan) and/or water dilution. After the dispersion was conditioned by concentration adjustment, it was degassed using a

centrifugal mixer (ARV-310, Thinky Corp., Tokyo, Japan) at 1400 rpm for 3 min under vacuum and then at 1800 rpm for 7 min under ambient pressure.

2.3. Nanopaper Substrate

The concentrated dispersion (1.0 wt %) was then evenly cast on an acrylic plate with an applicator. Cellulose nanopapers with thicknesses of 25 ± 2 μm were obtained after subsequent oven drying (DVS402, Yamato, Tokyo, Japan) at 55 °C under RH of 25%. For the thin layer and multilayered capacitors, a 0.36 wt % cellulose nanofiber dispersion was cast and dried on a glass plate, and cellulose nanopapers with thicknesses of 8 ± 1 μm were obtained. The thickness of the nanopaper was measured by a micrometer and field emission scanning electron microscopy (SU8020, Hitachi High Technologies Corp., Japan) at an accelerating voltage of 5.0 kV and a working distance of 16.9 mm.

2.4. Nanopaper Capacitor

The 5 mm × 5 mm electrodes were fabricated by inkjet printing silver nanoink (CCI-300, Cabot Printing Electronics and Displays, USA) on both sides of 25-μm-thick nanopaper, PET (Tetron HPE-25, Teijin Film Solutions Limited, Japan), and polyimide (Kapton(R) 100H, E. I. du Pont de Nemours and Company, USA) films using an inkjet printer (Dimatix DMP 2850, Dimatix-Fujifilm Inc., USA). Printed electrodes were sintered by heat treatment at 150 °C for 30 minutes. To confirm formation of the thin dielectric layer, 0.01–0.75 wt % cellulose nanofiber dispersions were cast and dried on 5 mm × 5 mm silver electrodes formed on nanopaper substrates. To prepare the multilayered nanopaper capacitor, a 0.1 wt % cellulose nanofiber suspension was cast and dried on a 5 mm × 5 mm silver electrode prepared on a nanopaper substrate, another silver electrode was fabricated, and the procedure was repeated.

2.5. Nanopaper Wireless Transmitter

An oscillation circuit based on a Colpitts oscillation circuit was fabricated on a nanopaper substrate. First, conductive lines and electrodes for the top layer were fabricated on the top side of the nanopaper substrate. The conductive lines and electrodes for the coil and capacitors were fabricated with a screen printer (Air Suction Screen Printer, Taiyoseiki Co., Ltd., Japan) using binder-free silver paste (TEC-PA-010, InkTec Co., Ltd., Korea). The resisters were fabricated with an inkjet printer (Dimatix DMP 2850, Dimatix-Fujifilm Inc.) using silver nanoink (CCI-300, Cabot Printing Electronics and Displays). Printed lines and electrodes were sintered by heat treatment at 150 °C for 30 minutes. To focus on the effect of the humidity on the capacitors, chip resistors were used in some experiments instead of printed resisters. Second, conductive lines and electrodes for the bottom layer were fabricated on the bottom side of the nanopaper substrate, and a chip transistor for amplifying the signal was mounted and connected with silver paste.

2.6. Characterization of the Nanopaper Capacitors and Nanopaper Wireless Transmitter

The capacitance of the fabricated capacitor was measured with an LCR meter (LCR-6200, Texio Technology Corp., China) at 1 kHz and 1 V, and the humidity was adjusted using a constant temperature and humidity chamber (PR-2 KT, ESPEC Corp., Japan). The measurement was performed in a 0% RH environment by placing a desiccant (diphosphorus pentoxide) in a closed container. The wireless signal from the nanopaper wireless transmitter was received by a rod antenna connected to a spectrum analyzer (GSP-9330, Texio Technology Corp., China).

2.7. Degradation Test

For the degradation experiment, we used natural soil. The soil (3 L) was collected at 34° 49′ 31.4′ ′ N 135° 31′ 22.6′ ′ E in February 2019 and kept in a plastic box. The temperature and humidity of the soil were well controlled at 27 ± 2 ° C and 99%, respectively. As control experiments, wireless transmitters on polyimide and PET substrates were also investigated.

3. Results and Discussion

To minimize the environmental impact, the nanopaper sensor device was fabricated by taking into consideration both the manufacturing process and materials. The nanopaper sensor device is shown in Figure 1a. It has three functions: sensing, wireless transmission, and degradability. The three passive elements (the resistor, coil, and capacitor) are mounted on nanopaper derived from natural wood. Ink-jet and screen-printing circuit fabrication techniques called printed electronics are attracting attention as eco-friendly processes because they do not require hazardous chemicals and high energy consuming facilities such as vacuum vapor depositing equipment. [22–24] Nanopaper is suitable for printed electronics because of its surface smoothness, printability, and heat resistance. Fine pitch conductive lines with line width/spacing (L/S) of 150 μm/150 μm were fabricated on the nanopaper substrate (Figure 1c), and sufficient levels of precision for fabricating a resistor, a coil, and a capacitor are expected. The most important element of this sensor device is the capacitor, which is an essential element of the wireless transmitter and humidity sensor. In this device, nanopaper is both the dielectric material and substrate of the capacitors, thereby achieving both eco-friendliness and degradability, as well as sufficient performance for wireless transmission. The nanopaper sensor device consists of a wood-derived nanopaper substrate and dielectric layer, silver lines and electrodes for the coil, resistors,

capacitors, and a Si chip transistor for signal amplification. That is, the device is made from plant material with minimal metals and minerals.

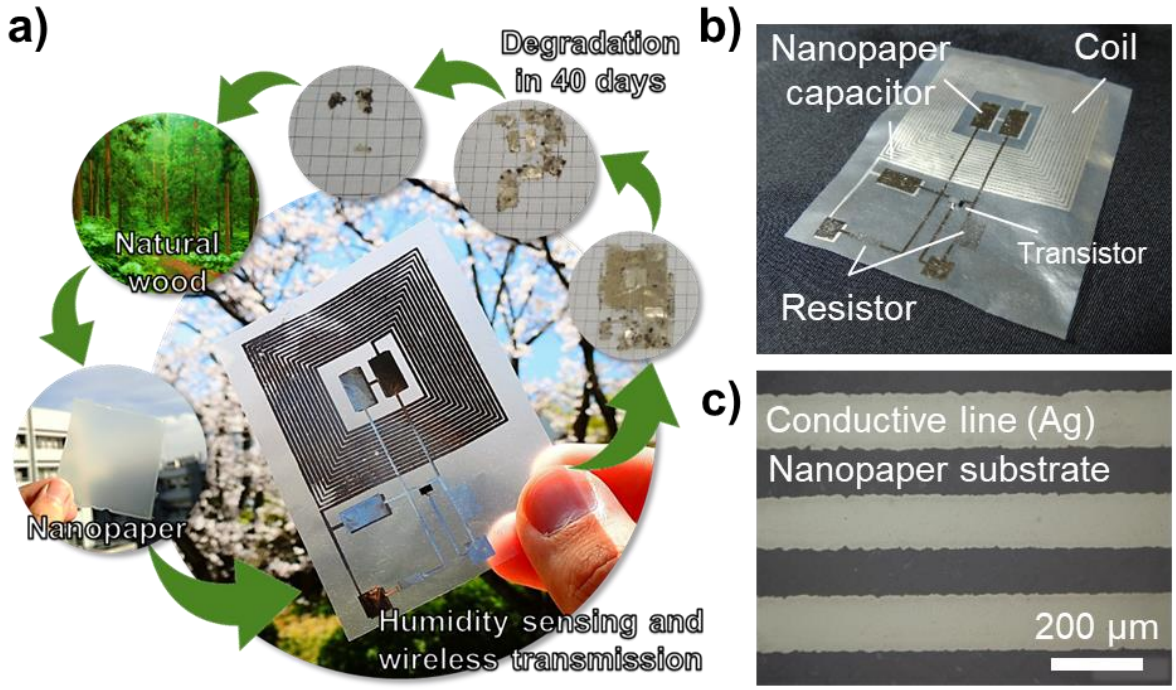

Figure 1. a) Nanopaper sensor device derived from natural wood. First, nanopaper is prepared from cellulose nanofibers extracted from natural wood. Conductive lines and electrodes are then fabricated on the nanopaper by inkjet and screen printing processes. b) Three passive elements (resistor, coil, and capacitor) mounted on nanopaper. c) Fine-pitch (L/S 150 μm/150 μm) conductive lines on the nanopaper substrate. The high heat resistance and surface smoothness of the nanopaper play important roles in fabricating a highly conductive substrate. The nanopaper sensor device can detect the change in the humidity and transmit the information by wireless transmission owing to the properties of the nanopaper as a dielectric layer for capacitors. After 40 days, more than 95% of the total volume of the device degrades and is returned to the soil.

The capacitor is an essential element of wireless transmitters. To prepare capacitors with sufficient capacitance (C [F]) for wireless transmission, there are three important parameters: the relative permittivity (ε_r) of the dielectric material (ε_0 [F/m] : the dielectric constant of vacuum) and thickness (d [m]) of the dielectric layer and the electrode area (A [m^2]). The capacitance can be expressed as

$$C = \frac{\varepsilon_0 \varepsilon_r A}{d} \quad (1)$$

There is a need for "green" dielectric materials with high relative dielectric constant and insulating properties that can withstand thinning and be easily layered.

To evaluate the performance of the nanopaper as a capacitor dielectric layer, we compared its performance with other polymer materials.[25] Capacitors with dielectric layers of the same thickness (25 μm) were fabricated using nanopaper, polyimide, polyethylene terephthalate (PET), and pulp paper, and their capacitances were measured with an inductance, capacitance, and resistance (LCR) meter. The capacitance of the nanopaper capacitor was about 2.6 times higher than those of polyimide and PET (Figure 2a). Common polymer materials, such as engineering plastics (e.g., polyimide and PET) and biodegradable plastics (e.g., polylactic acid), have relative permittivity of about 3.[25–27] In contrast, nanopaper has a dielectric constant of 5–6.[16] The high capacitance of the nanopaper capacitor is considered to be because of the high relative dielectric constant of nanopaper. Pulp paper derived from wood, similar to nanopaper, has long been used as capacitor dielectric layers.[28] However, pulp paper cannot be used as a thin capacitor dielectric layer because there are a large number of holes between the top and bottom electrodes, resulting in weak electrical insulation (Figure 2b). Because the cellulose nanopaper is composed of cellulose nanofibers with widths of 3–15 nm, it is possible to obtain a smooth surface enough for electrode fabrication (Figure 2c).

Next, thin film formation of the nanopaper was evaluated. The nanopaper was found to be sufficiently insulating for capacitance measurements of dielectric layers with thicknesses of 200 nm to 12 μm, and the capacitance increased with decreasing thickness of the dielectric layer (Figure 2d). The capacitance inversely increased with increasing thickness of the dielectric layer,

as expected from Equation 1. We confirmed that the nanopaper could form a thin film of sufficient thickness for use as a capacitor dielectric layer.

Finally, multilayer formation of the nanopaper dielectric layer was investigated. A nanopaper substrate was first fabricated on a slide glass, and then electrode and nanopaper dielectric layers were repeatedly fabricated. The capacitance of the nanopaper layered capacitor linearly increased with increasing number of layers (Figure 2e). It is desirable to increase the electrode area to improve the capacitor performance (Equation 1). However, because the two-dimensional size is limited in mounting, the electrode area is generally increased three-dimensionally by laminating or winding. In stacking of the nanopaper capacitors, the nanopaper dielectric layers could be stacked without any special pretreatment (Figure 2f). In other words, the multilayered nanopaper capacitor can be fabricated by only a printing process with low environmental impact and it does not require a harmful solvent or a high-energy deposition apparatus, and sufficient performance is expected when it is used for radio transmission.

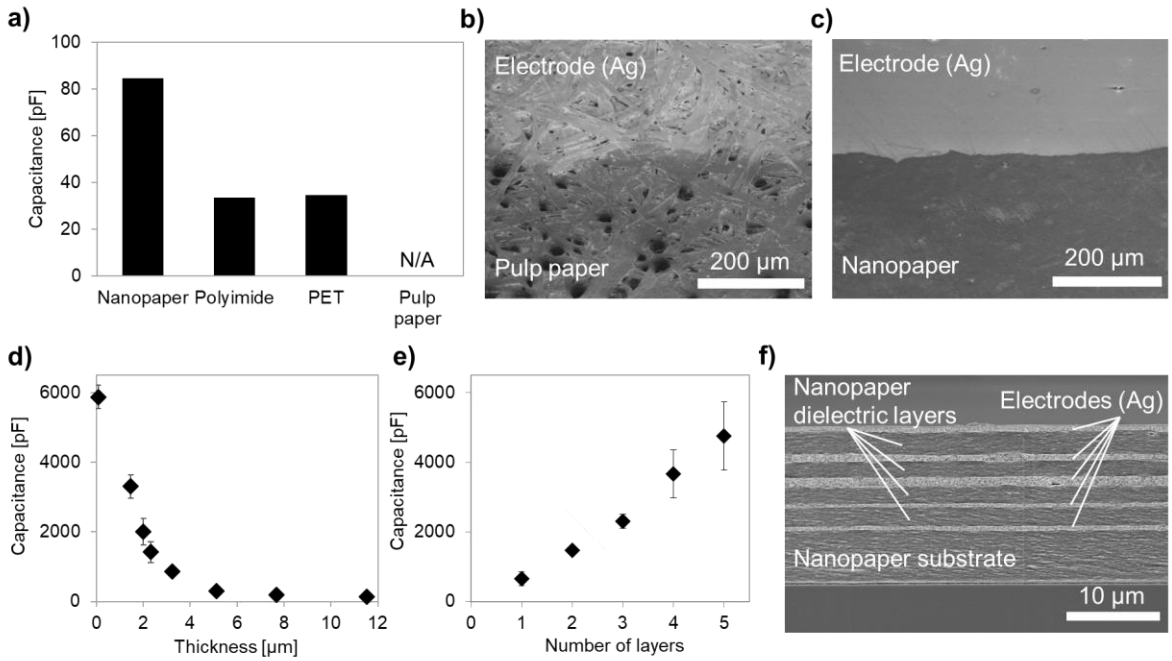

Figure 2. a) Capacitance of parallel-plate capacitors fabricated by mounting 25 mm × 25 mm electrodes on both sides of 25-μm-thick nanopaper, polyimide, PET, and pulp paper dielectric layers at 25 °C and 0% RH. The nanopaper capacitor has 2.6 times higher capacitance than the polyimide and PET capacitors because of the high relative dielectric constant and insulation between the electrodes. b) Densely packed structure of the nanopaper capacitor. c) Pulp paper (25 μm thick) capacitor. Because of holes between the pulp fibers, insulation between the electrodes could not be maintained and the capacitance could not be measured. d) Capacitance of nanopaper capacitors with 200-nm- to 12-μm-thick dielectric layers. e) Capacitance of multilayered nanopaper capacitors. f) Field-emission scanning electron microscopy image of the cross-section of the multilayered nanopaper capacitor.

By successfully fabricating a nanopaper capacitor with sufficient performance, it was possible to fabricate a wireless transmitter by printing and mounting the three main passive elements (capacitors, resisters, and coils) on a nanopaper substrate. Wireless transmission in the megahertz band was confirmed by measuring the signal from the nanopaper wireless transmitter with a spectrum analyzer (Figure 3a). Typically, electronic devices are fabricated by integrating various elements, wires, and substrates made of different materials, such as silver, copper, solder, FR-4, polyimide, and PET. Because heat treatment, such as sintering of conductive lines, soldering, and preliminary heating, is frequently performed during device fabrication, it is necessary to understand and control the differences in the heat resistance and linear thermal expansion coefficient of each element of the device. In particular, heat treatment at 100 °C or higher is often required in the process of wiring fabrication or soldering.[24] To use a biodegradable plastic, such as polylactic acid or polyvinyl alcohol, for a substrate or a dielectric layer, special contrivance, such as transfer printing, is required.[8] In fabrication of the nanopaper wireless transmitter, the substrate and dielectric layer were both made of high heat resistant nanopaper, and no effect was observed even after heat treatment at 150 °C for 30 min. Further, because the same material is used for the substrate and dielectric layer, problems such as peeling owing to a difference in the coefficient of linear thermal expansion did not occur.

The environmental performance of the nanopaper wireless transmitter circuit was evaluated for environmental monitoring applications. The device was installed outdoors, and the change of the oscillation peak with time was observed. The results are shown in Figure 4b. The oscillation frequency changed over time. The change of the signal was compared with meteorological information to clarify which parameters of the outdoor environment affected the oscillation frequency. We found that the oscillation frequency followed the change of the relative humidity (RH, Figure 3b). This means that the nanopaper wireless transmitter already functioned as a sensor device with humidity sensing and wireless transmission functions.

The humidity sensing mechanism was investigated. The circuit diagram of the fabricated oscillation circuit is shown in Figure 3c. The theoretical oscillation frequency can be expressed as

$$1/f = 2\pi\sqrt{L\frac{C_1C_2}{C_1+C_2}} \quad (2)$$

In the measurement environment of the nanopaper wireless transmitter, it is unlikely that the induction coefficient (L) of the coil changed, and it is considered that the capacitance of the nanopaper capacitor changed because of the influence of the humidity. Capacitors with nanopaper, polyimide, and PET dielectric layers were placed in an atmosphere of 25 °C and 20–80% RH, and the capacitance was measured (Figure 3d). The nanopaper capacitor greatly increased with increasing humidity. The capacitance of the capacitors with polyimide and PET dielectric layers hardly changed. It is known that the physical properties of nanopaper, such as the size, strength, and gas barrier properties, are easily affected by the humidity.[29-30] The moisture content of the nanopaper varied from 2.5% to 14.3% under 20–80% RH at 25 °C. The change in the capacitance is considered to be caused by the change in the moisture content in the

nanopaper. The capacitance of the nanopaper capacitor increased by up to about 3.6 times according to the humidity. Owing to the large change in the capacitance, it is considered that a basic oscillation circuit could output the change in the humidity. These results show that the capacitor with nanopaper as the dielectric layer can be applied as a humidity sensor, and a wireless transmission circuit with the nanopaper capacitor can be used as a humidity sensor device.

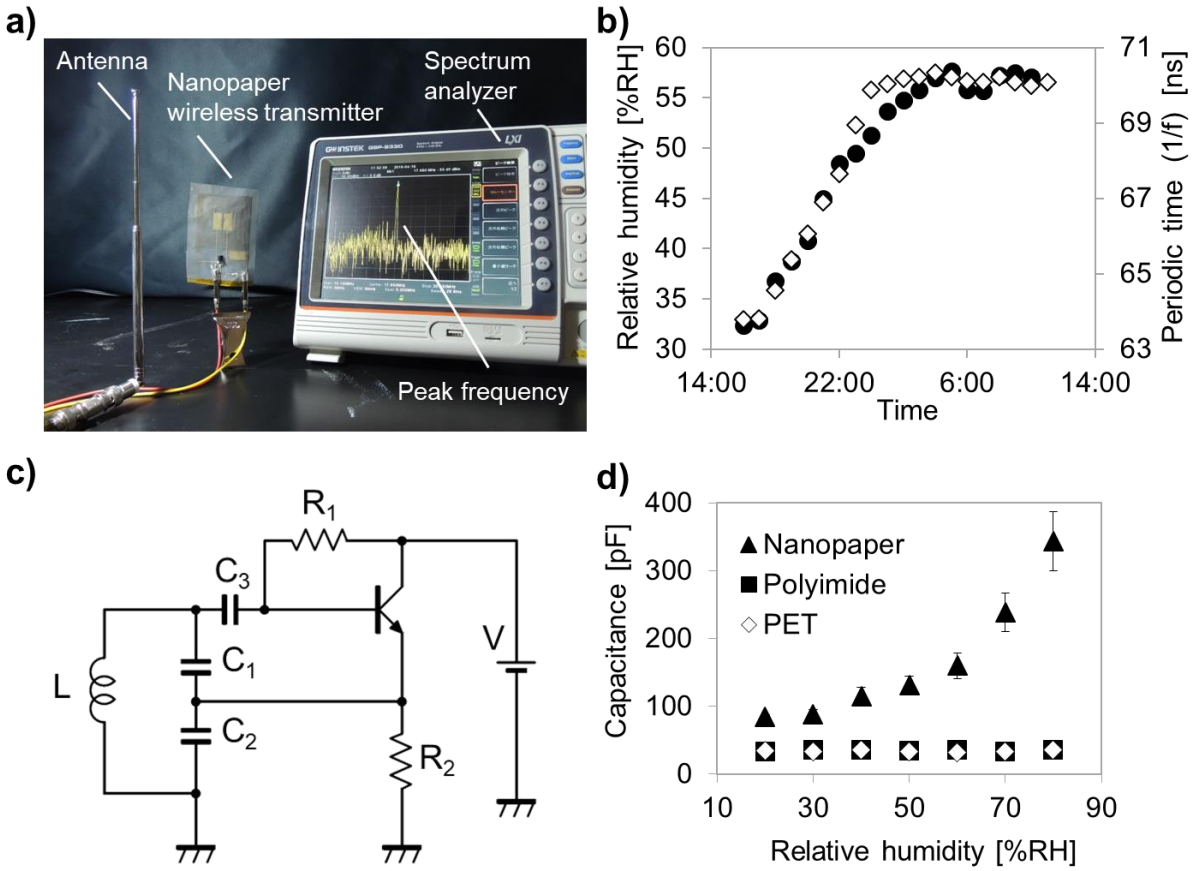

Figure 3. a) Megahertz band radio emission from a nanopaper wireless transmitter detected by a rod antenna connected to a spectrum analyzer. b) Change of the peak frequency of the nanopaper wireless transmitter with the RH. c) Simplified circuit diagram of the nanopaper wireless transmitter. This circuit is based on the Colpitts oscillator and the change in the oscillation peak is mainly caused by the changes in L, C_1, and C_2 (Equation 2). Because it is unlikely that the induction coefficient (L) of the coil would significantly change in this measurement environment, it is considered that the capacitance of the nanopaper capacitor changed. d)

Capacitance of the nanopaper capacitor at 25 °C and 20–80% RH, showing that the capacitance greatly increases with increasing humidity.

To confirm the degradability of the nanopaper sensor device, a degradation test in soil was performed. More than 95% of the total volume of the nanopaper device degraded in 40 days (Figure 4a). In contrast, the polyimide and PET substrates were stable after 40 days without any degradation. On day 10, biodegradation of the substrate part of the nanopaper sensor device was visually confirmed. After day 10, degradation of the substrate mainly occurred, and about 50% of the total volume was decomposed on day 20. There was a delay in decomposition around the conductive line part because of the antibacterial effect of silver. However, on day 30, degradation was progressing in the part where the wiring originally existed. This is considered to be because the silver wiring was gradually peeled off by the effect of swelling of the nanopaper substrate owing to moisture and decomposition proceeded from this part. By day 40, more than 95% of the total volume was degraded and lost. The substrate biodegraded and the conductive lines were degraded to levels indistinguishable from soil particles. From the above results, the fabricated sensor device has the three functions of sensing, information transmission, and degradability.

Realization of degradable sensor devices makes it possible to realize a wide range of future sensor devices (Figure 4b, c, d). The nanopaper sensor device can be mass-produced using only an eco-friendly process and resources. Because almost all of sensor device components decompose and do not need to be collected, the sensor devices can be used in large quantities. For example, it is possible to install a large number of weather observation devices in places such as streets and utility poles without worrying about management and disposal, which provides a very powerful way of accurate weather prediction and capturing local weather

changes, such as torrential rain and tornadoes. In addition, spraying humidity sensor devices and gathering information on large farms can improve the efficiency of water supply for crops. Information can be collected and managed for each crop, making it a powerful way of increasing food production and decreasing food loss. This method is more flexible than conventional methods for monitoring the natural environment, and it has great potential for applications such as automation of forestry by managing the growth of individual trees, environmental prediction, and maintenance and management of ecosystems. Other possible applications of nanopaper sensor devices include congestion prediction through highly accurate traffic information collection, efficient operation of small-scale power generation facilities, such as photovoltaic and wind power generation facilities, energy management in factories and manufacturing plants, and disaster prediction, typified by earthquakes and eruptions. The nanopaper sensor devices are expected to not only replace existing IoT devices, but also to significantly contribute to the development of new observation methods that take advantage of the no collection feature.

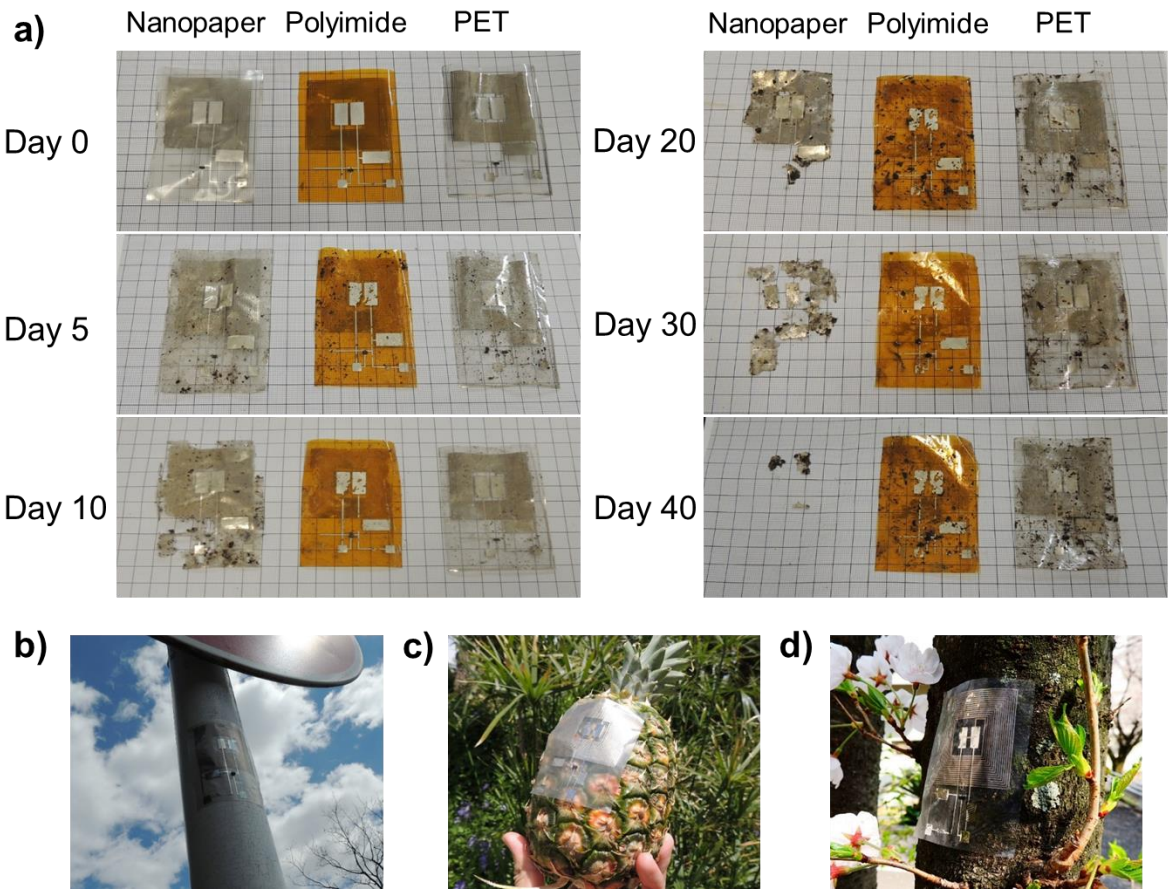

Figure 4. a) Time series of photographs of the nanopaper, polyimide, and PET sensor devices after burying them in soil collected at 34°49′31.4″N 135°31′22.6″E in February 2019. The humidity in the test environment was controlled at 99% RH. The nanopaper sensor device significantly degraded over time, whereas the other substrates were completely intact after 40 days. Examples of the potential uses of nanopaper sensor devices. b) Formation of a hyperdense meteorological observation network by attaching nanopaper sensor devices to electric poles, road signs, and so forth to collect weather information. c) Increase of food production by application to smart agriculture, and reduction of food loss by real-time monitoring of the storage conditions of packages, such as agricultural products. d) Efficiency improvement of forestry by monitoring the growth of individual trees, environmental forecasting, and measurement.

4. Conclusion

In conclusion, we have successfully fabricated a nanopaper sensor device with the three functions of sensing, information transmission, and degradability using nanopaper derived from natural wood. Because of its high dielectric constant, thin film formability, and lamination

properties, nanopaper shows high suitability for the dielectric layer of capacitors, which are essential for information transmission. A wireless transmission circuit was constructed using a nanopaper substrate and dielectric layer by an eco-friendly printing process, and it functions as a humidity sensing device owing to the moisture sensitivity of nanopaper. More than 95% of the total volume of the nanopaper sensor device decomposed in soil in 40 days. Nanopaper sensor devices will contribute to reduce environmental pollution and management and decrease the disposal costs of sensor devices, which are bottlenecks in development of an IoT society. They also demonstrate the basic concept of next-generation sensor devices.

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Author Contributions

Takaaki Kasuga and Masaya Nogi designed this work and prepared the manuscript. Takaaki Kasuga planned and performed experiments. Takaaki Kasuga, Hitomi Yagyu, Hirotaka Koga, Kojiro Uetani, and Masaya Nogi analyzed the results and discussed the manuscript during its preparation. All authors discussed the results and implications and commented on the manuscript at all stages.

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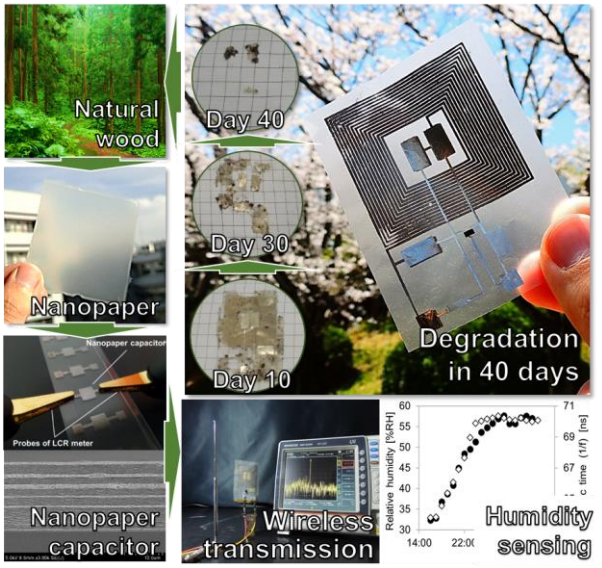
TOC


Natural wood
Nanopaper
Nanopaper capacitor
Probes of LCR meter
Nanopaper capacitor
Day 40
Day 30
Day 10
Degradation in 40 days
Wireless transmission
Relative humidity [%RH]
60
55
50
45
40
35
30
14:00
71
69
67
time (1/f) [ns]
Humidity sensing